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Physics Procedia 2 (2009) 1449–1453

**Physics
Procedia**www.elsevier.com/locate/procedia

Proceedings of the JMSM 2008 Conference

Preparation and characterization of macroporous ceramic supports for membranes

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Received 1 January 2009; received in revised form 31 July 2009; accepted 31 August 2009

Abstract

The objectives of this work were to prepare the ceramic supports from clays. These raw materials have been dictated by their natural abundance (low price) and their beneficial properties. The powders mixed with certain organic additives have been extruded to fabricate a porous tubular configuration with highly uniform porous structures. Subsequently, the influence of the sintering temperature on the total porosity, average pore size, pore size distribution and strength of supports is investigated.

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PACS: 61.43.Gt; 81.20.Hy; 81.05.Je

Keywords: Supports; Membranes; Porosity; ceramic

1. Introduction

Ceramic membranes have a large potential over their polymer counterparts for applications at high temperatures, pressure and in aggressive environments [1–4]. Generally, porous ceramics supports are needed for membranes manufacturing. In fact, the top layer (membrane) is closely related to its support [5]. In addition, the quality of the support is of crucial importance to the integrity of the membrane layers that are applied in the subsequent preparation steps. The surface roughness and homogeneity of the support will determine the integrity of these membrane layers, and, the surface roughness determines the minimal thickness of the membrane layer for complete surface coverage. These Tubular-type kaolin supports which were prepared by extrusion method are destined to be used as supports of micro-filtration membranes. These supports permit to provide mechanical strength to a membrane top layer to withstand the stress induced by the pressure difference applied over the entire membrane and must simultaneously have a low resistance to the filtrate flow.

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2. Experimental procedure

The chemical composition of the clays used in the present work given in weight percentages of oxides is: 50.56 wt% SiO_2 ; 34.15 wt% Al_2O_3 ; 1.15 wt% Fe_2O_3 ; 0.02 wt% CaO ; 0.31 wt% MgO ; 0.28 wt% TiO_2 ; 7.18 wt% K_2O ; and a 6.35 wt% of solids lost by calcination. The 2 main preparation processes, used in this work, are described in Fig.1. The tubular support was obtained by extrusion of the mixture of kaolin (80 wt %) and starch (20 wt%) as an organic additive. The flat configuration of supports was obtained using roll pressing technique and this configuration was used for mechanical tests. Finally, the total porosity, average pore size and pore size distribution have been determined by mercury intrusion porosimetry for supports sintered at different temperatures for 60 min.

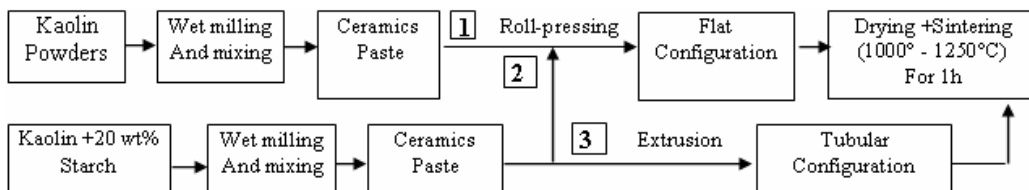


Fig. 1. A schematic diagram showing the main processes (1, 2 and 3), used for membrane supports preparation, in this work.

3. Results and discussion:

Structural evolution of the powder using Thermo Gravimetric (TG) and Differential Scanning Calorimetry (DSC) analysis shows the weight loss ratio of kaolin. These Two analyses have been carried out under air. The heating rate of the compacts from room temperature to 1200°C was 10°C/min.

TG curves, recorded during compacts heating (figure 2), permit the following remarks. A total weight loss ratio of about 8% of kaolin compacts is measured. In fact, this weight loss consists of two distinct stages. The first one is attributed to the humidity, whereas the second stage is related to water departure (by vaporization) existing in the kaolin chemical composition itself. The weight loss ratio of the last stage is more pronounced (Fig. 2).

These observations are also confirmed by DSC analysis (figure 2). Moreover, the first and third endothermic phenomena appeared at 65° and 497°C, respectively. They correspond to the water departure, whereas the second peak (at 251°C) corresponds to the departure of organic materials. Another stage which characterized by an exothermic reaction appeared at about 983°C. The origin of this reaction is not trenched yet. Some workers attribute this reaction to spinel formation while others attribute it to mullite nucleation.

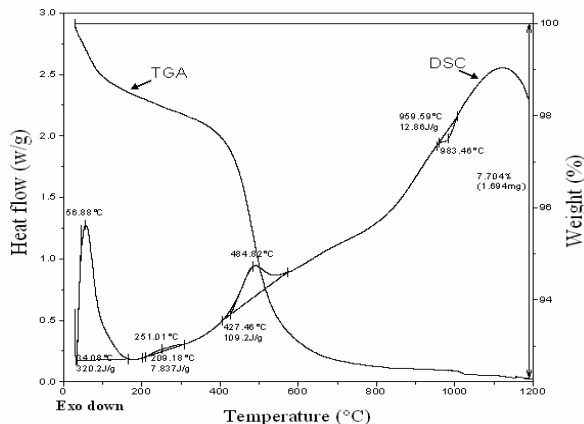


Fig. 2. Thermal Analysis (DSC+TGA)

The porosity measurement and the average pore size have been carried out for supports sintered at different temperatures for 60 minutes. The obtained results are illustrated in figures 3 and 4. As would be expected, these figures show, generally, that there is an increase in average pore size and a decrease in total porosity in samples, when the sintering temperature is increased.

Moreover, it can be said that both the average pore size and porous volume are closely related to the preparation method. The obtained results (figures 3 and 4) show that the starch addition to kaolin has a positive effect on the porosity ratio of supports compared to those prepared from kaolin alone. For example, the kaolin (K) support had a porosity ratio of $\approx 27\%$ and an average pore size around $0.5 \mu\text{m}$, whereas the kaolin + 20 wt% starch (K+S) supports had a porosity ratio of $\approx 46\%$ and an average pore size around $1.4 \mu\text{m}$, for samples sintered under the same conditions (at 1200°C for 1 hour). Consider figure 5 which presents a modal of pore size distribution, for samples sintered at 1200°C for 1 hour, it is almost Single (mono) Modal of Pore Size Distribution or homogenous pore distribution. This is necessary for a good integrity of the membrane. Fig. 6 shows the scanning electron microscope images of the cross section of the ceramic tube sintered at 1200°C . The morphology of the section suggests that the absence of macro defects and a good pore size distribution are a key condition leading to a good quality supports.

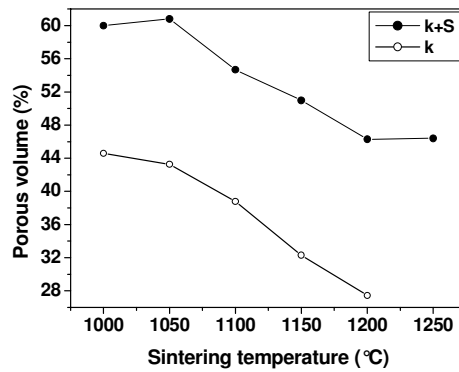


Fig. 3. Porous volume (%) versus sintering temperature for kaolin (k) and kaolin +20% starch (k+s) samples. using process 1 and 3.

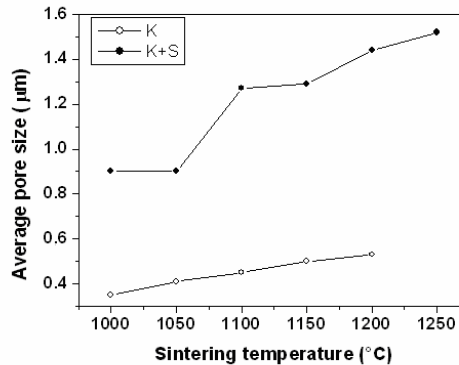


Fig. 4. Average pore size versus sintering temperature for kaolin (k) and kaolin +20wt% starch (k+S) samples.

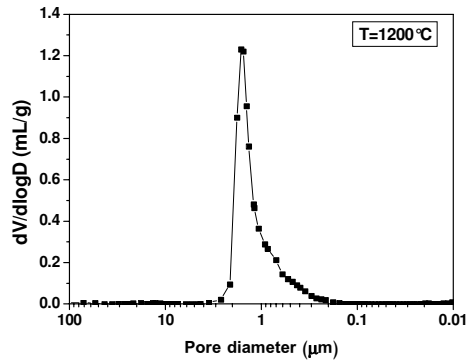


Fig. 5. Pore size distribution in kaolin+ 20 wt% starch samples sintered at 1200°C for 1 hour.

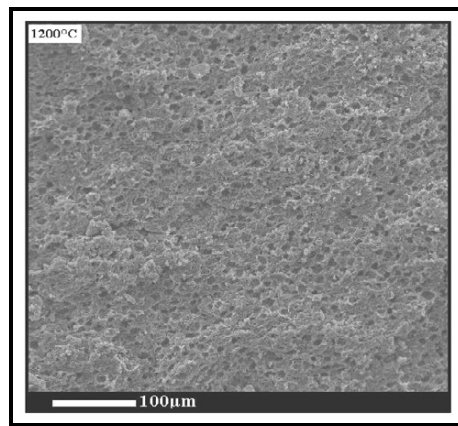


Fig. 6. SEM micrographs of sample sintered at 1200°C.

The flexural strength of porous ceramics (having different porosity ratios) was evaluated. The effect of sintering temperature on the flexural strength was also investigated. These porous kaolin samples have different microstructures, in terms of porosity. Their mean pore diameters and porosities were from 0.35 to 0.53 μm and 27% to 44%, respectively. As shown in Fig. 7, the flexural strength values of the porous kaolin samples without addition were higher than those of porous kaolin + organic additive samples. This figure shows that the flexural strength is closely related to the total porosity ratio which in its turn sintering temperature-dependant.

For example, flexural strength was 38 MPa at a porosity of 27.5% and an average pore size around 0.5 μm , whereas flexural strength was about 18 MPa for k+S supports having a porosity ratio of \approx 46% and an average pore size around 1.4 μm . Both K and K+S supports were sintered at 1200°C for 1 hour.

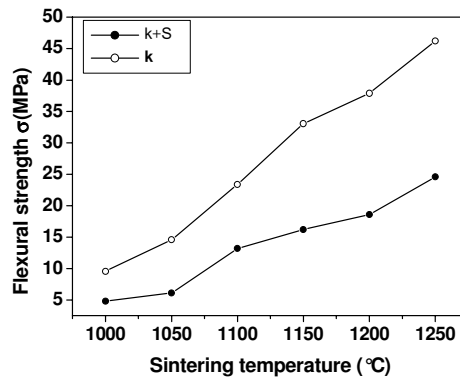


Fig. 7. Flexural strength as a function of sintering temperature for kaolin (k) and kaolin + 20 wt% starch (k+S) samples, using process 1 and 2.

4. Conclusions

In this work, ceramic membrane supports were prepared by extrusion method. Furthermore, the effect of temperature on porosity, flexural strength and average pore size of supports was investigated. The obtained results enable to conclude that clay supports can be used as a support for micro filtration and ultra filtration membranes. Moreover, these supports are characterised by a reduced manufacture cost since the used raw materials are very abundant (in Algeria) and their mechanical properties seem to be acceptable.

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